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# Evaluation of Commercial RO Membrane for $BaCl_2$ Separation

S. V. Huliienko & V. V. Yasenchuk

Faculty of Chemical Engineering, National Technical University of Ukraine "Igor Sikorsky Kyiv Polytechnic Institute", 37 Peremohy Ave., 03056 Kyiv, Ukraine

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## ABSTRACT

The conventional methods of the $BaCl_2$ solution separation are characterized by high energy consumption, reagent requirement, and other technological limitations. The pressure-driven membrane process can be an alternative, but the number of published investigations in this field is limited. The study of the effectivity of the $BaCl_2$ diluted solution separation using the commercially available reverse osmosis membrane was carried out. It was defined that the spiral wound membrane module HID TFC 1812-75 GPD (nominal permeate flux is 281 l/d) has acceptable characteristics of productivity and selectivity. In particular, the permeate flux through these membranes for the $BaCl_2$ solutions does not differ from the fluxes for other salt solutions in the considered range of the applied pressures (0.2-0.6 MPa). The rejection coefficient values were in the range of 0.9-0.95 and slightly varied with applied pressure. Therefore, such membranes are suitable for wastewater purification from the remains of $BaCl_2$. At the same time, the concentration ratio does not exceed 1.5-2, consequently, the application of such membranes on the concentration stage requires deeper justification based on the economical parameters. The significant influence of the concentration polarization on the separation effectivity was not detected up to applied pressures 0.5 MPa.

*Keywords*: Barium chloride, reverse osmosis, membrane, concentration, rejection

## 1.0 INTRODUCTION

Barium Chloride is commonly used as a cleaning agent in some chemicals productions and lubrication additive to oils. It is also used as raw material or intermediate product in the production of molecular sieves, chemical reagents, pigments, and coating for paper, and as an ingredient in salt mixtures for thermal treatment [1].

Except for the common application, this compound is widely used as a reagent for wastewater treatment [2-5]. In particular, $BaCl_2$ is the effective reagent for the removal of sulfate-ions [2] and radium [4-5]. Because this Barium Chloride is applied for mine water treatment in coal [3, 5] and Uranium mining [4] and also for wastewater treatment in pigments production [2]. In recent years, Barium Chloride is used as an absorbent in thermochemical refrigeration cycles [6-8]. Also, the novel applications include microcapsule production [9] and nanoparticle synthesis [10].

$BaCl_2$ is produced from barite ore (the main component is Barium Sulfate), the technological process includes disintegration, mixing with petroleum coke, and reduction into Barium Sulfide under high temperatures. Then, Barium Sulfide is purified and dissolved in water. The Barium Sulfide solution is treated with Hydrochloric Acid, and the Sulfide Acid is removed into a gas form, whereas the $BaCl_2$ remains in the solution. The solution is concentrated by evaporation and crystalized [1]. This method requires using a significant amount of expensive reagents, some of which are toxic and corrosive [1]. In

* Corresponding to: S. V. Huliienko (email: sergii.guliienko@gmail.com)
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addition, it was pointed out that Barium Chloride is a toxic compound that can cause diseases of the cardiovascular system [11-13].

Taking into account these factors, during $BaCl_2$ manufacturing special attention should be paid to the questions of separation, considering both the final product removal based on economic considerations and the wastewater treatment based on environmental reasons. As it was mentioned above, in the technological process of Barium Chloride production for separation such processes as evaporation and crystallization are applied. In particular, the processes of the crystallization from the subcritical state of liquid were proposed [14], also the ion exchange process can be used [1]. For the effluent treatment, the process of reagent precipitation is applied. The essence of this process lies in the treatment of the solution by Sulfate Acid or soluble sulfates with low-soluble Barium Sulfate formation [12-18]. The application of the adsorption process for such purposes also was proposed [19].

The disadvantage of the evaporation process is the high energy consumption, and the reagent precipitation does not allow the Barium Chloride reuse. Therefore, the question of more effective separation of $BaCl_2$ aqueous solution is still actual. It is well known that pressure driven membrane processes, including reverse osmosis, are highly effective for the separation of diluted salt solution in water [20, 21]. However, the question of applying such processes for Barium Chloride is not described in the literature with satisfying fullness. In most cases, the separation of $BaCl_2$ is considered during multicomponent solutions treatment. Moreover, in some works, the influence of the $Ba^{2+}$ ions on the membrane fouling was pointed out during the separation of multicomponent solutions by reverse osmosis [22-26]. In particular, these investigations were carried out for the cases of brackish [22-23] and seawater desalination [25]. The high risk of $BaSO_4$ precipitation in such cases is underlined [22-26]. Also, the formation of the antifouling mineralized coating with Barium ions was investigated [27]. Some publications were dedicated to the separation of multicomponent solutions containing Barium Chloride by nanofiltration [28-30]. In this works, the effectiveness of $Ba^{2+}$ ion removal is considered more comprehensively. However, in work [28], it was pointed out that the selectivity of nanofiltration membrane for $Ba^{2+}$ is lower than other bivalent cations. The investigations of separation solutions containing $BaCl_2$ by electrodialysis were also carried out [31-32]. In these works, Barium Chloride was used for the examination of the transport and selectivity properties of cation exchange membranes. The transport of $Ba^{2+}$ with liquid membranes was considered in the work [33]. However, the available published data do not allow us to conclude about the effectivity of the Barium Chloride separation by membrane processes and the suitability of these processes for the modernization of Barium Chloride manufacturing plants.

Taking into account the conclusion about the selectivity of nanofiltration membranes for $Ba^{+2}$ reported in work [28], it is reasonable to consider the application of reverse osmosis membranes for Barium Chloride solution separation. The aim of this work is the examination of the effectivity of aqueous Barium Chloride solution with using of commercially available reverse osmosis membranes and the determination of the rationality of the application of reverse osmosis in the technological line of $BaCl_2$ production.

## 2.0 METHODS

### 2.1 Materials

For the preparation of the experimental solution, the crystalline $BaCl_2$ purchased from TM Kleberg (Rivne, Ukraine) and reverse osmosis permeate obtained in the laboratory of the National Technical University of Ukraine "Igor Sikorsky Kyiv Polytechnic Institute" were used.

The experiments were carried out for spiral wound membrane modules of the lower price segment HID TFC 1812-75 GPD purchased from Green Life Co., Ltd. (China) and manufactured by HID Membrane Co., Ltd. (China) [34]. The manufacturing declares the following characteristics for used membrane modules [34]:

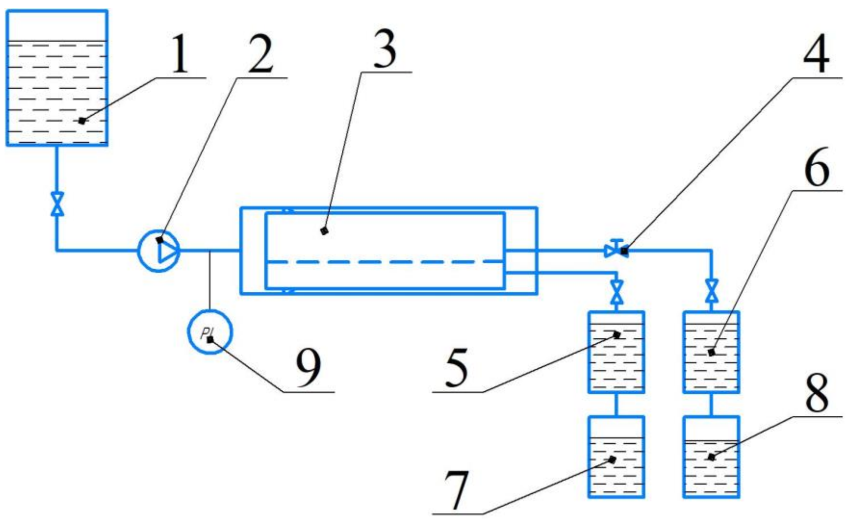
- Stable salt rejection is 97%;
- Minimum salt rejection is 96%;
- Average permeate flow is 380 l/d (11.7 l/h);
- Effective area of the membrane is 0.38 $m^2$.

These characteristics were obtained for the testing solution of NaCl 500 ppm with the applied testing pressure of 0.45 MPa and the testing temperature of 25°C [34].

### 2.2 Experimental set-up

The experiments were carried out on the existing set-up, which was used in a previous investigation [35, 36]. The set-up includes the feed solution tank 1, pump 2, membrane apparatus 3, the permeate control tank 5, the retentate control tank 6, the permeate storage tank 7, the retentate storage tank 8, the pressure gauge 9, and the regulation valve 4 [35].

The set-up allows us to carry out the measurements of the permeate and retentate flow rates by the volumetric technique (the direct measurements include measurement of volume with accuracy ±5 ml and time with accuracy ±0.01 s). The concentration in the feed solution, permeate, and retentate was measured by the portative TDS-meter with an accuracy of ±0.01 ppm. The applied pressure was measured by pressure gauge with an accuracy of ±0.01 MPa, and the temperature of the feed solution was measured by the block of the Chromel-Copel thermocouples (not shown on the scheme) with an accuracy of ±0.2°C [36].




1 – feed solution tank; 2 – pump; 3 – membrane apparatus; 4 – regulation valve; 6, 7 – control tanks; 7, 8 – storage tanks; 9 – pressure gauge

**Figure 1** The scheme of the experimental set-up [35]

For the solution preparation, the samples of the crystalline Barium Chloride were made, the mass of which was measured by the electronic scale with an accuracy of ±0.1 g.

### 2.3 Experimental Procedure and Main Measurements

In the current work the same experimental procedure as in work [35] was used. Tank 1 was filled with the prepared solution of $BaCl_2$ with a predetermined concentration, the predetermined value of the applied pressure was established by the regulation valve 4, and the obtained during the separation process permeate and retentate simultaneously collected in the control tanks 5 and 6 during the predetermined time [35].

The productivity was evaluated by the values of the transmembrane flux ($l/(h \cdot m^2)$), which was determined by the following dependence [35]:

$$J = \frac{L_p}{A} \quad (1)$$

where $L_p$ is the permeate flow rate, l/h; $A$ is the membrane surface area, $m^2$.

The selectivity was evaluated by the rejection coefficient value [37]:

$$R = 1 - \frac{c_p}{c_F} \quad (2)$$

where $c_p$ is the concentration of $BaCl_2$ in permeate; $c_f$ is the concentration of the $BaCl_2$ in the feed solution.

Also, for the evaluation of the concentration effectiveness, the ratio of concentration in retentate and feed solution called the concentration ratio was used:

$$K = \frac{c_r}{c_f} \quad (3)$$

The results of experiments were compared with the Kimura-Surirajan model. According to it, the transmembrane flux is proportional to the applied pressure [37, 38]:

$$J = A_p\left(\Delta p - \Delta \pi\right) \quad (4)$$

where $A_p$ is the permeation constant; $\Delta p$ is the applied pressure; $\Delta\pi$ is the osmotic pressure difference.

For the processing of the experimental results, the least square method was applied.

## 3.0 RESULTS AND DISCUSSION

The experimental investigations were carried out for the concentration range 100, 200, 400, and 600 mg/l, the applied pressure was varied from 0.2 to 0.6 MPa. The measurements were done under the environmental temperatures without heating or cooling the feed solution. The temperature of the feed solution was varied in a range of 14-16°C. In this case, the influence of the temperature on the separation process can be reasonably assumed as negligible. The considered range of applied pressure includes the testing value used by the manufacturer for the membrane characterization [34], however, the experimental temperature was lower than the testing value.

The transmembrane flux as a function of the applied pressure is shown in Figure 2. As it was expected, the flux values increase with increasing applied pressure and decreasing of the feed solution. Such dependence is approximated linearly with good enough accuracy. In particular, the values of the squares of the selective correlation coefficient $R^2$ were 0.9813, 0.9875, 0.9762, and 0.977 for concentrations 100, 200, 400, and 600 mg/l correspondingly. These values of the $R^2$ are close to values used in work [39] for verification of model parameters. This allows us to suggest that the experimental results are in agreement with the well-known theoretical statement (Kimura-Surirajan model). This indicates that during the separation of the $BaCl_2$ solution by reverse osmosis, the special effects which complicate the process are absent. The biggest deviation from linear dependence was observed for the applied pressure value 0.6 MPa, moreover, in the decreasing direction. These results can be explained by the influence of concentration polarization, which is a typical phenomenon for pressure driven membrane processes [40]. Also, such effect was observed during the separation of the others diluted salt solutions [36].

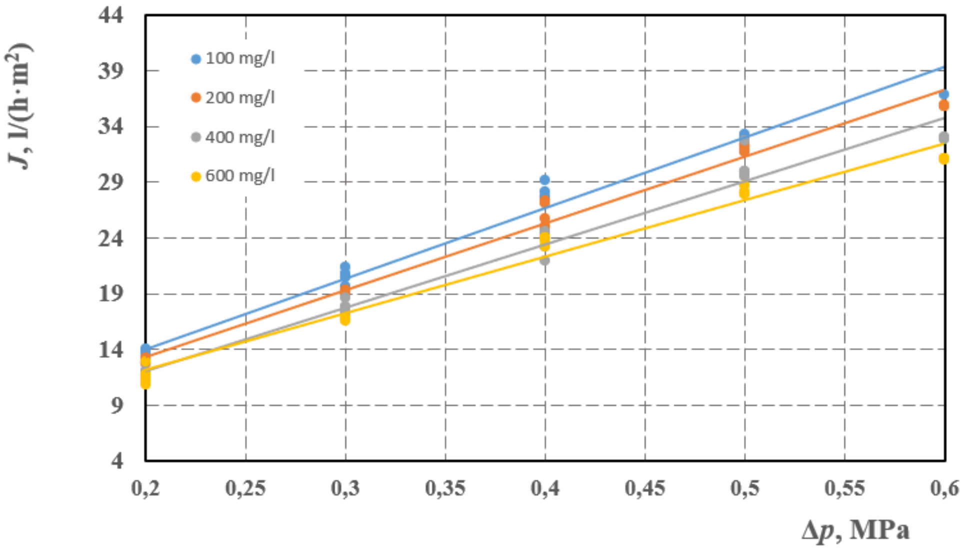



**Figure 2** The dependence of the transmembrane flux from applied pressure

The experimentally obtained value of average permeate flux for the testing value of applied pressure provided by the membrane manufacturer was 26.48 l/(h·m$^2$). At the same time, according to the data provided by manufactured data, for NaCl solution corresponding value is 27.38 l/(h·m2), which is higher by 3.28%. However, the experimental data was obtained for lower temperature value and, consequently, for higher solvent viscosity [41], which decreases membrane flux according to [40].

The values of flux are also on the same level that was observed during the application of the considered types of membrane modules for other salt solutions, in particular, during the separation of the $MgSO_4$ and $NaNO_3$ solutions in work [36]. Therefore, by the productivity, the reverse osmosis process with the application of the commercially available membrane of lower price segment seems to be acceptable for the separation of the $BaCl_2$ solutions.

The characteristics of selectivity are represented in Figures 3 and 4. The value of the rejection coefficient variated in the range of 0.9-0.95 (Figure 3), and the significant influence of the applied pressure was not observed. Similarly, the noticeable impact of the feed concentration was not pointed out. These values are lower than the value provided by the manufacturer for testing solution [34], however, in previous work [36] for the other salt solutions the salt rejection was in the same range (90-95%).

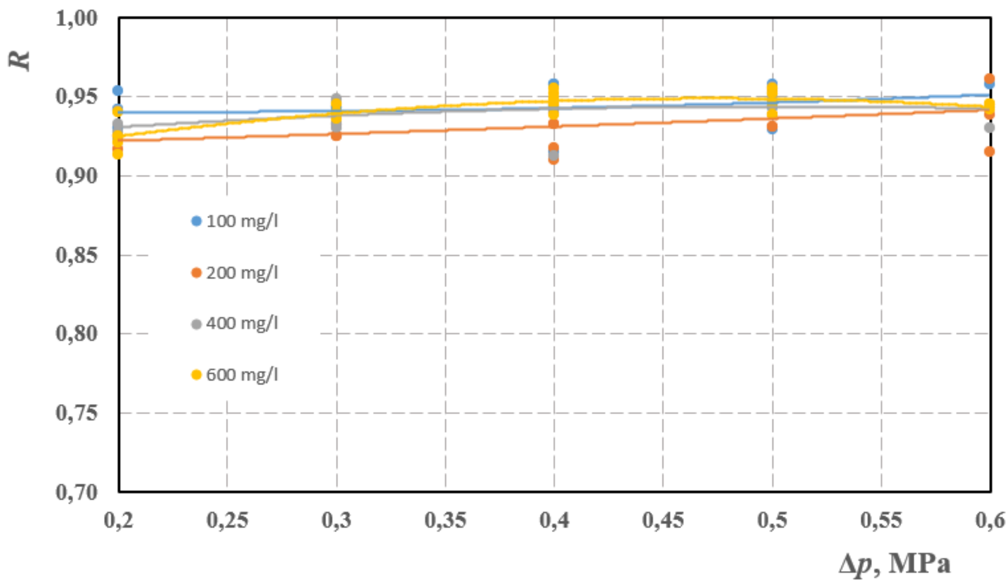



**Figure 3** The dependence of the rejection coefficient from applied pressure

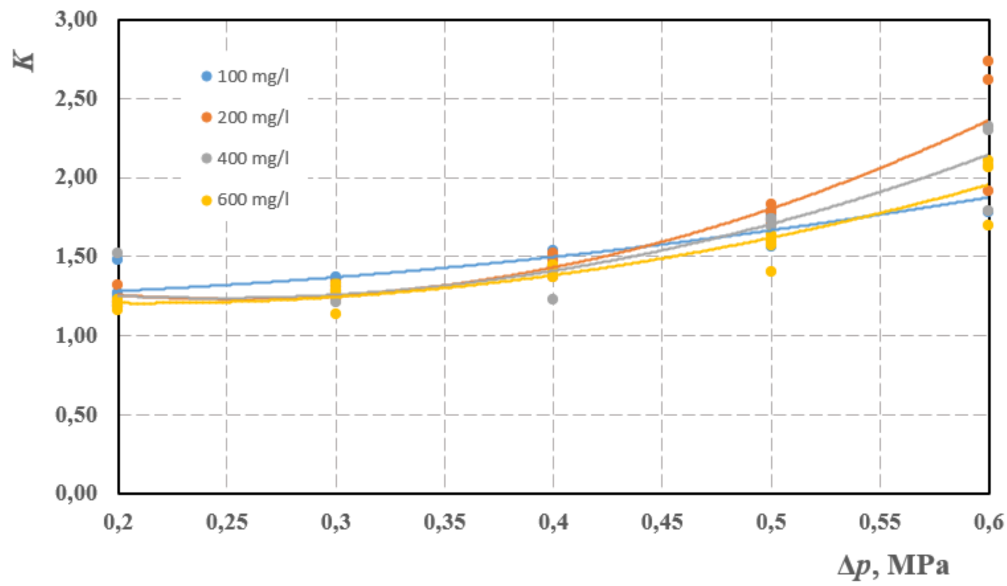



**Figure 4** The dependence of the concentration ratio from applied pressure

At the same time, the values of the concentration ratio were moderately increased with applied pressure (Figure 4). However, the values of this parameter were relatively low. In an addition, the ratio of the retentate and permeate fluxes dramatically decreased with applied pressure (Figure 5).

Significant fouling during experiments was not detected. This indicates that substantial fouling reported in works [22-26] is caused mainly by presence of sulfate ions. The $Ba(OH)_2$ is also characterized by relatively low solubility [41], and the risk of this compound precipitation also could be expected. However, the formation of Barium Hydroxide fouling layer during $BaCl_2$ was not observed under experimental conditions.

Such results indicate that the considered membrane modules are quite suitable for water purification from $BaCl_2$, and can be recommended for application in wastewater treatment systems for Barium Chloride removal.

However, the relatively low values of the concertation ratio complicate the use of the considering membrane type as the concentration technology in the Barium Chloride production line, at least with an application of the single-pass scheme, which was realized in the experimental set-up. For this purpose, in reasonably carrying out the detailed energy and economic analysis. The scheme with recirculation as in work [42] also can be considered.

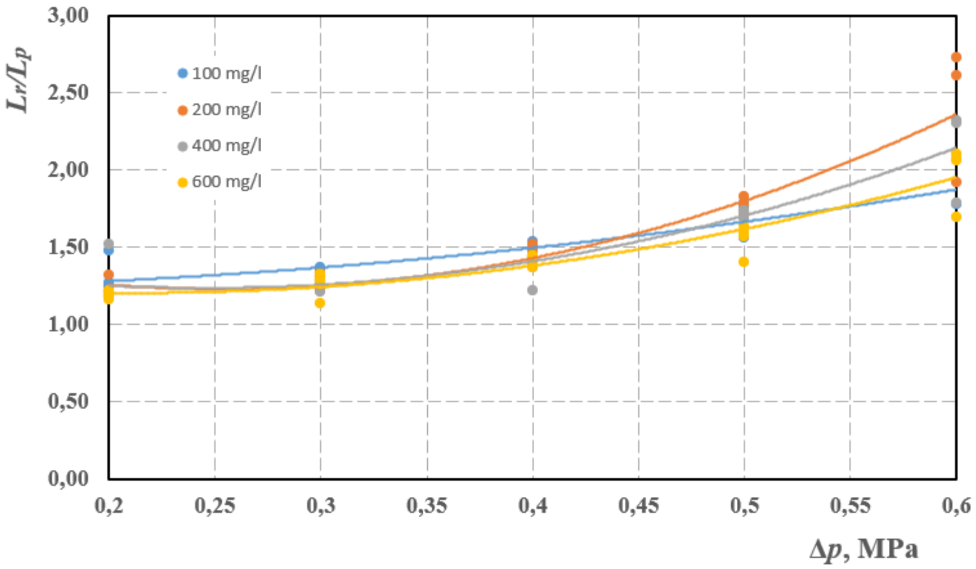



**Figure 5** The dependence of the ratio of retentate and permeate fluxes from applied pressure

The experimental results show that the most rational value of the applied pressure in the experimental condition is 0.5 MPa, since in that case the high value of transmembrane flux (28-33 l/(h·m$^2$)) and high value of the rejection coefficient (0.93-0.95) are observed, and also the significant influence of the concentration polarization is absent.

## 4.0 CONCLUSION

The commercially available reverse osmosis membranes of lower price segment show acceptable characteristics for the Barium Chloride solution separation. In particular, such membranes allow the removal of 90-95% $BaCl_2$ from the solution, which makes them suitable for purification systems. At the same time, the application of these membranes for the concentration stage requires deeper substantiation based on the economical parameters. The significant influence of the concentration polarization on the productivity and separation effectivity for the pressure values up to 0.5 MPa was not detected. Also, the other effects which influenced the reverse osmosis separation of such solution were not found. The most rational value of the applied pressure in the experimental condition is 0.5 MPa since in that case the high value of transmembrane flux (28-33 l/(h·m$^2$)) and high value of the rejection coefficient (0.93-0.95). The process of $BaC_2$ separation by reverse osmosis is in agreement with the Kimura-Surirajan model.

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